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78

30. Dez. 2004

To the
European Patent Office

Notice of Opposition to a European Patent

Tabulation Marks |

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I. Patent opposed		Opp. No. OPPO (1)
		Patent No. 1022115B1
		Application No. 00300413.2
		Date of mention of the grant in the European Patent Bulletin (Art. 97(4), 99(1) EPC) 31.3.2004
Title of the invention: Polymeric Articles Having a Textured Surface and Frosted Appearance		
II. first named in the patent specification	Atofina	
Proprietor of the Patent		
Opponent's or representative's reference (max. 15 spaces)		RWF/CK/M12621
		OREF
III. Opponent	OPPO (2)	
Name	LUCITE INTERNATIONAL UK LIMITED	
Address	1 st Floor Offices Queens Gate 15-17 Queens Terrace Southampton. SO14 3BP	
State of residence or of principle place of business	United Kingdom	
Telephone/Telex/Fax		
Multiple opponents	<input type="checkbox"/> further opponents see additional sheet	
IV. Authorisation		
1. Representative	Richard Frith OPPO (9)	
(Name only one representative to whom notification is to be made)		
Name	Appleyard Lees	
Address of place of business	15 Clare Road Halifax West Yorkshire HX1 2HY UK	
Telephone/Telex/Fax	441422330110	441422330090
Additional representative(s)	<input checked="" type="checkbox"/> (on additional sheet/see authorisation) OPPO (5)	
2. Employee(s) of the opponent authorised for these opposition proceedings under art. 133(3) EPC	Name(s):	
Authorisation(s)	<input type="checkbox"/> not considered necessary	
To 1./2.	<input type="checkbox"/> has/have been registered under No. 	

V. Opposition is filed against:

- the patent as a whole
- claim(s) No(s).

VI. Grounds for opposition:**Opposition is based on the following grounds:**

(a) the subject-matter of the European patent opposed is not patentable (Art. 100(a) EPC) because:

- it is not new (Art. 52(1); 54 EPC)
- it does not involve an inventive step (Art. 52(1); 56 EPC)
- patentability is excluded on other grounds, i.e.

(b) the patent opposed does not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Art. 100(b) EPC; see Art. 83 EPC).

(c) the subject-matter of the patent opposed extends beyond the content of the application/ of the earlier application as filed (Art. 100(c) EPC, see Art. 123(2) EPC).

VII. Facts and arguments

(Rule 55(c) EPC)
presented in support of the opposition are submitted herewith on a separate sheet (annex 1)

VIII. Other requests:

We request the patent is revoked in its entirety. If the Opposition Division does not accede to this request then we request Oral Proceedings (Article 116 (EPC)).

IX. Evidence presented		for EPO use only
<input checked="" type="checkbox"/> Enclosed = <input checked="" type="checkbox"/> <input type="checkbox"/> will be filed at a later date = <input type="checkbox"/>		
A. Publications:		Publication date
1 JP 04-2769668		
Particular relevance (page, column, line, fig.): 		
2 JP 59-038253		
Particular relevance (page, column, line, fig.): 		
3 JP 61-159440		
Particular relevance (page, column, line, fig.): 		
4 US 4,876,311		
Particular relevance (page, column, line, fig.): 		
5 US 5,395,822		
Particular relevance (page, column, line, fig.): 		
6 US 3,345,434		
Particular relevance (page, column, line, fig.): 		
7 US 5,063,259		
Particular relevance (page, column, line, fig.): 		
<input checked="" type="checkbox"/> Continued on additional sheet		<input checked="" type="checkbox"/>
B. Other evidence		
<input type="checkbox"/> Continued on additional sheet		<input type="checkbox"/>

CONTINUATION SHEET

IX Evidence Presented

A. Publications:

8. US 3,992,486
9. GB 2,220,002
10. US 5,621,028
11. The Physics of Glassy Polymers, 1997, Chapter 8 § 8.1 Rubber Toughening
12. Polymer Blends, 1999, p157, 170 and 173
13. US 4,000,216

X. Payment of the opposition fee is made

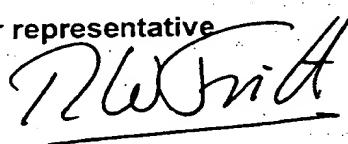
as indicated in the enclosed voucher for payment of fees and costs (EPO Form 1010)

**XI. List of documents:**

Enclosure No.:

No. of copies

0	<input checked="" type="checkbox"/> Form for notice of opposition	2	(min. 2)
1	<input checked="" type="checkbox"/> facts and arguments (see VII.)	2	(min. 2)
2	Copies of documents presented as evidence (see IX.)		
2a	<input checked="" type="checkbox"/> — Publications	2	(min. 2 of each)
2b	<input type="checkbox"/> — Other documents		(min. 2 of each)
3	<input type="checkbox"/> Signed authorisation(s) (see IV.)		
4	<input checked="" type="checkbox"/> Voucher of payment of fees and costs (see X.)	1	
5	<input type="checkbox"/> Cheque		
6	<input type="checkbox"/> Additional sheet(s))		(min. 2 of each)
7	<input type="checkbox"/> Other (please specify here):		

**XII. Signature
of opponent or representative**

Place Halifax, UK

Date 24 December 2004

Please print name under signature. In the case of legal persons, the position which the person signing holds within the company should also be printed.

Continuation of Box 2 (Representative).**Additional Representatives:**

APPLETON, Ben
BRANDON, Paul Laurence
BRIERLEY, Anthony Paul
CHUGG, David John
DAVIES, Robert Ean
FRITH, Richard William
JACKSON, Nicholas Andrew
KOHLER, Janet Wendy
MOY, David
NEILL, Alastair William
PIDGEON, Robert John
ROBINSON, Ian Michael
SHERWARD-SMITH, Hugh
WADDINGTON, Richard
WALSH, David Patrick

All of:

APPLEYARD LEES
15 CLARE ROAD
GB-HALIFAX HX1 2HY

OPPOSITION STATEMENT

European Patent No.: 1022115B
European Appl. No.: 00300413.2
Proprietor: Atofina
Address: 92800 Puteaux (FR)

1. GROUNDS AND EXTENT OF OPPOSITION

- 1.1 EP 1022115B (the patent) is opposed for all contracting states in respect of all claims.
- 1.2 The patent is opposed under Articles 100(a), 100(b) and 100(c) EPC, and in particular the patent contains subject matter which extends beyond the content of the application as filed, lack of novelty and lack of inventive step.

2. REQUESTS

- 2.1 We request revocation of all claims of the patent, and oral proceedings (Art 116 EPC) if the Opposition Division is not inclined to accede to this request.

3. PRIOR ART

- D1 JP 04-279668 (Japanese and English language translation)
- D2 JP 59-038253 (Japanese and English language translation)
- D3 JP 61-159440 (English language translation)
- D4 US 4,876,311
- D5 US 5,395,822
- D6 US 3,345,434
- D7 US 5,063,259
- D8 US 3,992,486
- D9 GB 2,220,002
- D10 US 5,621,028
- D11 The Physics of Glassy Polymers, 1997, Chapter 8 §8.1 Rubber Toughening
- D12 Polymer Blends, 1999, p157, 170 and 173
- D13 US 4,000,216

4. FIELD OF THE PATENT

4.1 The patent relates to an extruded polymeric article having a frosted and surface textured finish (claims 1 to 6), an extrudable resin (claims 7 to 11) and use thereof for forming an article having a frosted and surface textured finish (claim 13), use of a polymeric matrix and polymeric particles as defined in any one of claims 1 to 11 for preparing an article having a frosted and surface textured finish (claim 12) and the use of an article as defined in any one of claims 1 to 6 or a resin of claims 7 to 11 for preparing parts for lighting, furniture applications etc (claim 14).

4.2 The extruded polymeric article as defined in claim 1 has the following characteristics:

- (a) a polymeric matrix comprising an acrylic polymer;
- (b) substantially spherical, highly crosslinked polymeric particles comprising 10 to 50% styrene, 90 to 50% methyl methacrylate and 0.1 to 2.5% crosslinking agent;
- (c) the particles have a mean particle size of 25 to 55 micrometers and a particle size distribution between 10 to 110 micrometers.
- (d) the article has a surface textured finish;
- (e) the article has a frosted finish and the frosted appearance is achieved through the mismatch of the refractive indices of the polymeric particles and polymeric matrix by greater than 0.02.

We note that the claim includes the word "comprising" and thus not all of the polymeric particles must have the claimed mean particle size and particle size distribution, but only some of the particle present must include these features.

4.3 The extrudable resin as defined in claim 7 has the following characteristics:

- (a) 20 to 90% polymethyl methacrylate matrix;
- (b) 5 to 50% modifiers; and
- (c) 5 to 30% of highly crosslinked spherical beads comprising at least the same constituents as the beads as defined in claim 1 and having the same particle size and particle size distribution; with the exception that:

- (d) the mismatch of refractive indices of the highly crosslinked spherical beads and polymethyl methacrylate based matrix is greater than 0.2.

4.4 The extrudable resin as defined in claim 11 has the following characteristics:

- (a) 70 to 85% polymethyl methacrylate matrix;
- (b) 15 to 30% highly crosslinked spherical beads having the same particle size and particle size distribution of the beads as claimed in claim 1 and comprising 15 to 35% styrene, 65 to 85% methyl methacrylate and 0.5 to 1.5% allyl methacrylate;
- (c) the mismatch of refractive indices of the highly crosslinked spherical beads and polymethyl methacrylate based matrix is greater than 0.2.

5. **PROBLEM ADDRESSED BY THE PATENT**

The purpose of the patent is to produce a resin that may be used for extrusion applications to create a product with a surface texture and frosted (i.e. translucent) appearance thereby reducing the need for traditional surface treatments and adding organic fillers to the polymer matrix (paragraphs 0003 and 0004). It is noted however that the claims are not limited to exclude such organic fillers or where appropriate such surface treatments. The extruded polymeric article is particularly suitable for parts of lighting, signs, points of purchase or cosmetic displays, containers, home or office decorations, furniture applications, show doors or office doors, notably it may exhibit desired light transmittance and diffusion characteristics while having the required translucent appearance.

The problem is solved by the inclusion of a highly crosslinked polymeric spherical particle in an acrylic matrix. The frosted appearance of the thermoplastic composition is achieved through the mismatch of the refractive indexes of greater than 0.02 of the fine particles and the acrylic matrix (paragraphs 001 and 0032). Such a difference in refractive indices is achieved by employing a particle having a different chemical composition than the matrix. The surface texture is allegedly controlled by the degree of crosslinking and mean size of the particles (paragraph 0011). However, it is also clear from the patent that the "textured surface" can be generated by processing the polymeric composition through conventional extrusion equipment (paragraph 0044).

6. ADDED SUBJECT MATTER ARTICLE 123(2) EPC

We submit that claims 7, 11, 13 and claims 12 and 14 in so far as they refer to claims 7 to 11 offend Article 123(2) EPC. Thus, these claims and those claims dependent thereon (i.e. claims 8 to 10) are not allowable.

In particular, independent claims 7 and 11 both include the feature of:

"wherein there is a mismatch of refractive indices of the highly crosslinked spherical beads and polymethyl methacrylate based matrix by greater than 0.2"

However, the application as filed states that the difference in refractive index of the crosslinked particles and the matrix material should be greater than 0.02 (paragraphs 0011, 0034 and claim 5 as filed).

Moreover, if the patentee argues that this is an obvious error, which we submit is not the case, and it attempts to invoke Rule 88 (EPC). Then, we would argue that the proposed correction (i.e. a difference in refractive index of greater than 0.02) widens the scope of protection of the granted claims and therefore offends Article 123(3) EPC. Moreover, Rule 88 must be bound by Article 123 (EPC) as Article 164(2) EPC stipulates that an Article has precedence over a Rule (see G3/89).

7. LACK OF NOVELTY

7.1 Claim 1

7.1.1. In view of D1 we submit that claim 1 lacks novelty in view of D1.

D1 discloses a light diffusing resin comprising a resin having fine crosslinked polymer particles dispersed therein. The particles have an average particle size in the range of 1 to 30 μm with the standard deviation of the particle size distribution being 20% or less of the average particle size (i.e. a range of 0.8 to 36 μm) (see abstract, claim 1 and paragraph 0005). Thus, D1 discloses

particles having a size and particle size distribution which falls within the ranges as claimed (see §4.2 point (c)).

The resin of D1 may comprise a methacrylic resin (see claim 2 and paragraph 0010). In this respect, Example 1 employs a methyl methacrylate/ethyl acrylate copolymer (91:9). Thus feature (a) of §4.2 is disclosed by D1.

When a methacrylic resin is employed then the refractive index of the particles is 1.50 to 1.54, since the refractive index of methacrylic resin is typically 1.49 (paragraph 0012). Thus the mismatch of the refractive index of the particles and the resin is in the range 0.01 to 0.05 i.e. greater than 0.02. Thus, the finished article formed from the composition of D1 will have a frosted appearance as this is what claim 1 states. Hence, D1 discloses the features of §4.2 (e).

When methacrylic resin is employed as the base then the particles comprise a crosslinked copolymer of methyl methacrylate and/or styrene (paragraph 0009). In this respect, Example 1 which employs an acrylic resin includes crosslinked polymeric particles comprising 79 parts methyl methacrylate, 19 parts styrene and 2 parts crosslinker (ethylene glycol dimethacrylate). The particles are preferably spherical (paragraph 0007) and as they include an amount of crosslinking agent (2%) which falls within the claimed range, they are by definition "highly crosslinked". Thus D1 discloses the features of §4.2 (b).

The matrix resin and the polymeric particles may be compounded and pressed (i.e. extruded). Thus, the resin of D1 may be extruded into a polymeric article and by definition from the patent such an article will have a surface textured finish, because the polymeric particles of D1 comprise the same components as those claimed in the patent, they have the desired degree of crosslinking and mean particle size (paragraph 0011 of the patent) and the article of D1 is formed by extrusion (paragraph 0044 of the patent). Furthermore, it is common knowledge (see D4 col. 6, lines 18 to 23, D2, page 3 lines 10 to 18 and 28 to 37) that high crosslinked polymeric particles of the type disclosed in D1 maintain their shape during extrusion thereby producing a surface textured finish. Moreover, when the composition of D1 is formed into a film, the film may be screen printed (paragraph 0011). From the patent it is clear screen printing introduces surface texture (paragraph 0004 lines 19 to 20). Thus, D1 discloses features of §4.2 (d).

Hence, D1 discloses all of the features of claim 1.

7.1.2 In view of D4, we submit claim 1 lacks novelty in view of D4.

D4 relates to opaque synthetic resins which may be formed into articles, such as light projection screens, by extrusion (col 1, lines 8 to 11 and col 2, lines 49 to 51).

The synthetic resin comprises a polymer matrix (PM) such as an acrylic resin (col. 6, lines 1 to 8 and Example 2). Thus, D4 discloses the features of claim 1 as detailed at § 4.2 (a).

Dispersed within the polymer matrix are crosslinked spherical polymeric beads (col. 2 lines 56 to 61 and col. 4, lines 38 to 45) comprising 9.9 to 59.9% by wt of monomer A, such as styrene (col. 2, lines 63 to 65; col. 3, line 32 and Example 1), 90 to 40% of vinyl monomer B such as methyl methacrylate (col. 2, lines 66 to 68, col. 4, lines 8 to 10 and Example 1) and 0.1 to 20% by weight of a crosslinker (col. 3, lines 1 to 2 and Example 1). Thus, D4 discloses the features of claim 1 as detailed at § 4.2(b).

The difference of the refractive index between the particles and the matrix is at least 0.01, preferably 0.02 and more preferably 0.04 (col. 5, lines 64 to 67). Thus, D4 discloses the features of claim 1 as detailed at § 4.2(e) and as defined in the claim the article will have a frosted appearance.

The average size of the polymeric beads is from 5 to 50 microns, and preferably 20 to 35 microns (col. 3, lines 5 to 7) and during preparation of the polymeric beads "the size of the beads can be adjusted within the claimed range between 20 and 50 microns" i.e. the particle size distribution is between 20 and 50 microns. Thus, D4 discloses the features of claim 1 as detailed at § 4.2(c).

Suitably, as the beads have the desired particle size and include the claimed amount of crosslinker so that they maintain their shape during

extrusion (see col. 6, lines 18 to 23), the resultant article will have a surface textured finish.

Thus D4 discloses all of the features of claim 1.

7.2 *Claim 2*

7.2.1 In view of D1 the features of claim 2 are also disclosed by D1.

The crosslinked polymeric particles may be present at 1 to 10 wt% (paragraph 0012), thus the methacrylic resin may be present at 90% by wt, which as stated in relation to claim 1 may be an alkyl methacrylate/alkyl acrylate copolymer (see Example 1).

7.2.2 In view of D4, the features of claim 2 are also disclosed by D4.

The acrylic polymer matrix of D4 may comprise a polymethyl methacrylate homopolymer or a methyl methacrylate/alkyl acrylate copolymer (col. 3, line 50 to col. 4 line 10, col. 6, lines 4 to 8, and Example 2). The polymer beads are present in an amount of 1 to 30%, preferably 1.5 to 20%, in particular 2 to 10% by weight relative to the acrylic polymer matrix (col. 2, lines 56 to 61). Thus the acrylic polymer matrix is present at 99 to 70%, preferably 98.5 to 80%, in particular 98 to 90% by weight relative to the polymer beads.

7.3 *Claim 3*

7.3.1 In view of D1.

The crosslinked polymeric particles may be present at 1 to 10 wt% in the article of D1 (paragraph 0012). Thus, D1 discloses the features of claim 3.

7.3.2 In view of D4.

The crosslinked polymer beads may be present in an amount of 1 to 30% by wt, preferably 1.5 to 20% by weight, in particular 2 to 10% by wt in the article of D4 (col. 2, lines 56 to 61). Thus D4 discloses the features of claim 3.

7.4 **Claim 4**

7.4.1 In view of D4.

D4 discloses all of the features of claim 4. In this respect, we refer to our comments regarding claims 1 to 3. In addition, the crosslinker of D4 may comprise an allyl ester of methacrylic acid i.e. allyl methacrylate (col. 4, lines 27 to 34).

7.5 **Claims 5 and 6**

7.5.1 In view of D1.

The crosslinking agent of D1 may include divinyl benzene and ethylene glycol dimethacrylate (paragraph 0009 in particular lines 8, 9 and 19). Thus D1 discloses the features of claims 5 and 6.

7.5.2 In view of D4.

The crosslinkers disclosed in D4 include divinyl benzene, glycol di(meth)acrylate, and allyl methacrylate (col. 4, lines 16 to 18 and 27 to 34). Thus, D4 discloses the features of claims 5 and 6.

7.6 **Claim 11**

Ignoring the feature of refractive index relating to added matter, D4 discloses all of the features of claim 11. Claim 11 relates to an extrudable resin as defined in claims 1 and 4 and we refer to our comments thereon.

7.7 **Claim 12**

As stated in relation to claim 1 above, the article of D1 will inherently have a frosted and surface textured appearance. In this respect, we note the articles of the specific Examples of D1 at page 9 have a haze value of greater than 90%, which as stated in the patent represents one measure of a frosted appearance (see paragraph 0083 of the patent).

The articles of D1 will also be surface textured as the composition of D1 includes identical components to the composition as claimed in claim 1 of the patent. Moreover, as stated in relation to claim 1, the light diffusing resin of D1 can be melt compounded and pressed (i.e. extruded) (paragraph 0011).

Moreover, when the composition of D1 is formed into a thin film, then it may be screen printed (paragraph 0011). From the patent it is clear that screen printing introduces surface texture (paragraph 0004 lines 19 to 20).

Thus D1 discloses all of the features of claim 12.

As stated in relation to claim 1, the polymeric matrix and particles of D4 may be used to form an article having a frosted and surface textured finish. Thus claim 12 also lacks novelty in view of D4.

7.8 **Claim 13**

Our comments with respect to claim 12 for D4 apply equally for the extrudable resin as claimed in claim 11. Thus, claim 13 lacks novelty in view of D4.

7.9 **Claim 14**

The articles of D1 may be used for lighting covers, displays and signs (abstract). The articles of D4 may be used for light projection screens (col. 1, lines 8 to 11). Thus, D1 and D4 disclose the features of claim 14.

8 LACK OF INVENTIVE STEP

8.1 General

It is well known to those skilled in the art, that thermoplastic materials which have been surface treated and/or include fine inorganic filler particles may be used for lighting parts. However, although such thermoplastic materials may exhibit satisfactory light diffusion characteristics, typically such materials have unsatisfactory light transmittance and the physical properties of the resin matrix, such as impact strength, may deteriorate (see para 0004 of the patent and D2, page 3, lines 20 to 26). Thus, no invention resides in recognising the problem to be solved.

The solution to this problem as proposed by the patent is to provide an acrylic matrix which includes a highly crosslinked polymeric spherical particle, such that the resultant extruded product has the desired frosted appearance (translucent appearance) and "surface texture".

In particular, the desired frosted appearance is achieved by employing a crosslinked polymeric particle which has a refractive index of less than or greater than 0.02 compared to the acrylic matrix. However, it is also well known to those skilled in the art, that a mismatch of refractive index of a polymeric particle and a matrix resin will produce the desired translucent/frosted effect, moreover the mismatch of refractive index typically is greater than 0.02 (see D1 page 6, para 0012, D4, col 1, lines 18 to 22 and col. 5, lines 58 to 68, D5, col. 3 lines 48 to 52, D6 col. 2, lines 55 to col. 3, lines 3 and D8, abstract). Furthermore, it is evident from D8, col 5, line 20 to 50 that the mismatch of refractive indices may be achieved by employing a polymeric particle having a different chemical composition than the polymer matrix.

Moreover, it is well known to those in the art that crosslinked polymeric particles do not typically distort during processing (i.e. extrusion) compared with non-crosslinked particles (D4, col. 6 lines 18 to 23, D8, col. 6, lines 2 to 6, and col. 8, lines 1 to 8, col. 12, lines 15 to 24 and Table 1). Suitably, articles formed from a polymer matrix and crosslinked polymeric particles typically having a textured surface which diffuses light (D2, page 3 lines 10 to 18 and lines 28 to 37, D6 col. 2, line 49 to 64).

Thus, the overall solution to the problem solved by the patent is also known to those skilled in the art.

8.2 **Claim 1**

8.2.1. In view of D1 alone or in combination with any other document.

In the unlikely event it is held claim 1 is novel over D1, then we respectfully reserve the right to argue that claim 1 is obvious in view of D1 alone and/or in combination with any other document.

In this respect, D1 lies in the same technical field as the patent, namely that of polymeric resins for lighting applications having satisfactory light transmittance, diffusion and haze. Hence, a skilled person would be aware of this document.

D1 also provides an identical solution to the same problem addressed by the patent, as D1 teaches a skilled person that the combination of a highly crosslinked styrene/methyl methacrylate spherical particle having the claimed particle size and particle size distribution in combination with an acrylic polymer matrix may be melt compounded and compressed to form an article having the desired light transmittance, diffusion and haze.

In respect of D1, the above comments apply equally to claims 2, 3, 5, 6, 12 and 13.

8.2.2. In view of D4 alone or in combination with any other document.

Similarly, in the unlikely event, it is held claim 1 is novel over D4, then we also respectfully reserve the right to argue that claim 1 is obvious in view of D4 alone and/or in combination with any other document.

D4, like D1, lies in the same technical field as the patent and provides an identical solution to the same technical problem addressed by the patent.

In respect of D4, the above apply equally for claims 2, 3, 4, 5, 6, 11, 12, 13 and 14.

8.2.3. In view of D2 alone or in combination with D4 and/or D1.

D2 lies in the same technical field as the patent and solves the same technical problem in an identical way by providing an acrylic resin molding having excellent light diffusion properties, higher light transmittance and satisfactory haze value (i.e. frosted appearance) (p2, lines 34, pg 3, paras 1 to 4 and p 11, line 18).

In particular, D2 discloses a light diffusing acrylic resin molding formed by an extrusion molding procedure for light covers (p 9, lines 21 to 31). The acrylic resin molding comprises an acrylic polymer matrix (100 parts) blended with 1 to 30 parts by weight of a crosslinked polymer (p4, lines 1 to 5).

The crosslinked polymer is formed in the same way as the polymeric particles of the patent (p8, final para) namely suspension polymerisation, and it is therefore spherical as evidenced by the term beads at p10, line 31. The spherical crosslinked polymer has a mean particle size of 10 to 500 microns, preferably 35 to 200 microns (col. 4 line 39), the lower limit at least falls within the claimed range.

The spherical crosslinked polymer comprises 0.5 to 5 pts wt of a crosslinkable monomer and 100 pts by wt of non-crosslinkable monomers (p4, lines 10 to 12) comprising 50 to 90% of a C₁ to C₄ alkyl methacrylate, preferably methyl methacrylate (p5, lines 23 to 27 and line 39) and 3 to 15% of an aromatic vinyl monomer such as styrene (p6, lines 13 to 16). Thus, the spherical crosslinked polymer of D2 includes the claimed amount of crosslinker and thus it would be regarded as "highly crosslinked". The crosslinked polymer of D2 includes the same chemical entities as the particulate of the claimed patent and it is added to an identical acrylic matrix. Thus, the difference in refractive index of the spherical crosslinked polymer and the acrylic matrix of D2 will be greater than 0.02 and an article formed therefrom would have a frosted appearance. In this respect, the acrylic resin molding disclosed at page

10, line 35 to page 11 line 21 has a haze value of 79 (p11 line 18). Suitably, the opposed patent states a high haze value is an indication of a frosted appearance (see para 0083).

D2 also discloses the light diffusion property of the article namely the surface texture, is not affected by secondary processing unlike prior art processes (page 3, lines 13 to 18 and lines 34 to 38). Thus the article of D2 has a surface textured finish. This is in line with the patent as the polymeric particles of D2 include the claimed amount of crosslinker, the claimed amounts of identical monomers, and the desired particle size (see para 0011 of the patent). Moreover, as evidenced by D4 and D8 in particular (see § 8.1) highly crosslinked polymeric particles do not typically distort during processing.

Thus D2, discloses all of the features of claim 1 apart from explicitly disclosing a particle size distribution of 10 to 110 microns.

Firstly, the patent does not disclose any advantage of using the claimed particle size distribution. Thus, no inventive step has been demonstrated by employing such a distribution.

Secondly, the preferred particle size as disclosed in D2 is 35 to 200 microns. Thus, if the skilled person operated within the whole range, or even at the lower limit of 35 microns, as this is what D2 teaches him to do, then it is inherent and a statistical certainty that at least some, if not the majority of particles will have a diameter that falls within the range or around the selected lower limit of 35 microns. Thus at least some of the particles of D2 will have the claimed particle size distribution of 10 to 100 microns. Hence, claim 1 lacks an inventive step in view of D2, as claim 1 of the patent does not specify that all of the particles must have the claimed particle size distribution, but only some of the particles must have this feature.

In addition, D2 states it is undesirable to employ polymer particles having a size of greater than 500 microns and less than 10 microns (page 5, para 1). Thus, a skilled person on reading D2 will realise the particle size distribution should be between 10 to 500 microns. Thus, at

least some of the particles would have the claimed lower limit of the particle size distribution as claimed and at least some of the particles would have the size distribution within the claimed range per se, particularly when operating D2 within the preferred range of particle sizes i.e. 35 to 200 microns.

D4 teaches a skilled person that the average particle size is from 5 to 50 microns, preferably 20 to 35 microns, and the particle size distribution is between 20 to 50 microns (col. 3, lines 5 to 7 and col. 5 lines 19 to 20). Thus a skilled person reading D4 and D2 together would employ particles in the method of D2 having a particle size at the lower limit of the range disclosed in D2, namely 10 to 50 microns, preferably 20 to 35 microns, with a particle size distribution of 20 to 50 microns, as this is what D4 teaches him to do. Hence claim 1 lacks an inventive step in view of D2 and D4.

D1 teaches a skilled person that the crosslinked polymer particles should have a narrow size distribution of 20% or less of the average diameter (claim 1 of D1). This is in accordance with D4. Thus a skilled person reading D1 and D2 together would employ such a particle size distribution, as this is what D1 teaches him to do, particularly as D2 also states it is undesirable to have a particle size of less than 10 microns and above 500 microns (page 5, para 1 of D2). Thus operating with the preferred range of average particle sizes of D2 (i.e. 35 to 200 microns), the particle size distribution would be 28 to 240 microns. Thus at least some of the particles would have the claimed particle size distribution of 10 to 110 microns. Thus, claim 1 lacks an inventive step in view of D1 and D2.

8.2.4 In view of D3 alone or in combination with D4 and/or D1.

D3 discloses a light scattering acrylic resin composition obtained by compounding 1 to 40 parts by weight of a crosslinked polymer with 100 parts by weight of methyl methacrylate polymer (p1, para 2).

The crosslinked polymer may be formed by suspension polymerisation (Embodiment 1) and has a particle size of 30 to 300 microns (page 1,

para 2) the lower limit of which falls within the claimed range. The crosslinked polymer comprises 0.3 to 3 parts crosslinking monomer per 100 parts of a non-crosslinking monomer comprising 10 to 90 wt% methyl methacrylate and 20 to 40 wt% styrene (page 1, para 2, page 2, last para and page 3, para 2).

Thus, within the definition of the patent the crosslinked polymer is regarded as "highly crosslinked" as it includes the claimed amount of crosslinker. Moreover, the crosslinked polymer of D3 includes the same chemical entities as the particulate as claimed in the patent and it is added to an identical acrylic matrix. Thus, the difference in the refractive index between the acrylic matrix and the crosslinked particle of D2 will be greater than 0.02 thereby producing a frosted appearance. This is endorsed by the disclosure at page 3, lines 13 to 16 where the haze value is greater than 85%, which we know from the patent is a measure of translucence (para 0083).

The light scattering acrylic resin is formed by compounding and it may be formed into articles by extrusion (page 1, para 2 and page 4, para 2). Thus, such an article will be surface textured as we know from D2, D8 and D4 (see § 8.1) that highly crosslinked particles do not deform during extrusion and the patent states that by employing the claimed particles during extrusion produces an article having a textured surface (para 0041 of the patent).

Thus, D3 discloses all of the features of claim 1 apart from explicitly stating the average particle size distribution of between 10 to 110 microns.

The preferred particle size as disclosed in D3 is 30 to 300 microns. Thus, if the skilled person operated within the whole range, or even at the lower limit of 30 microns, as this is what D3 teaches him to do, then it is inherent and a statistical certainty that at least some of the particles will have a diameter that falls within the range or around the lower limit of 30 microns. Thus at least some of the particles of D2 will have the claimed particle size distribution of 10 to 110 microns. Hence, claim 1 lacks an inventive step in view of D3 alone.

Claim 1 also lacks an inventive step in view of a combination of D3 and D4 as well as D3 and D1 for identical reasons given in respect of D2 (see § 8.2.3), and our comments apply equally.

8.3. *Claim 2*

8.3.1 In view of D2 alone or in combination with D1, D3 and/or D4.

The matrix of D2 comprises a methyl methacrylate homopolymer or a methyl methacrylate/alkyl acrylate copolymer (p9, para 2). The matrix is present at 100 parts by weight to 1 to 30 parts by weight crosslinked polymer. Thus, the matrix is present at 99 to 76.9% by wt and the crosslinked spherical beads at 1 to 23.1% by wt. Thus, D2 discloses all the features of claim 2.

The modifier in claim 2 represents an optional feature. D2 also discloses the resin may include such modifier (p9, lines 28 to 31). Moreover, the patent states that the inclusion of such modifier is well known in the art (para 0019) as evidenced also by D7, D11 and D12 (impact modifiers), D2 p9 lines 27 to 31; D10 col. 3 line 20 onwards and D6 col. 6 lines 4 to 19. Thus, no inventive step resides in this optional feature.

8.3.2 In view of D3 alone or in combination with D1, D2 and/or D4

The acrylic resin matrix of D3 comprises a methyl methacrylate homopolymer or a methyl methacrylate/alkyl acrylate copolymer (page 2, penultimate para). The composition of D3 includes 1 to 40 parts by weight crosslinked polymer to 100 parts by weight acrylic resin matrix e.g. 99% to 71% by wt acrylic matrix resin and 1 to 29% by wt crosslinked polymer (page 3, para 1). Thus, D3 discloses the features of claim 2.

8.4 *Claim 3*

8.4.1 In view of D2 alone or in combination with D1, D3 and/or D4.

Our comments at § 8.3.1 apply equally as well as the disclosure at p9, lines 11 to 14 of D2. Hence, D2 discloses the features of claim 3.

8.4.2. In view of D3 alone or in combination with D1, D2 and/or D4.

Our comments at §8.3.2 apply equally as well as the disclosure at p3, para 1 of D3. Hence, D3 discloses the features of claim 3.

8.5 **Claim 4**

8.5.1 In view of D2 alone or in combination with D1, D3 and/or D4.

The matrix of D2 may be polymethyl methacrylate and present in an amount of 76.9% to 99% by wt. Suitably, the crosslinked beads may be present in an amount of 1 to 23.1% by wt (see § 8.3.1 above). The beads may include 3 to 15 wt% styrene (p6 para 3), 55 to 90% by wt methyl methacrylate (page 5, line 35 to page 6 line 2), and 0.5 to 6 pts wt per 100 parts non-crosslinkable monomer of a crosslinker such as allyl methacrylate (page 7, line 6, and lines 14 to 17). Hence, D3 discloses the feature of claim 4.

8.5.2 In view of D3 alone or in combination with D1, D2 and/or D4.

We refer to our comments at §8.3.2 and §8.2.4. In addition, the acrylic glass preferably contains 10 to 30 parts by weight of the crosslinked polymer to 100 parts by weight of acrylic resin (p3, para 1). Thus the acrylic resin is present at 90% to 76.9% by weight and the crosslinked polymer 10% to 23.1% by weight. Moreover as evidenced by D2 (see §8.5.1 above) and embodiment 1 of D3, allyl methacrylate is regarded as a standard crosslinking agent and the patent does not exemplify any advantage by using this crosslinking agent.

8.6 **Claims 5 and 6**

Claims 5 and 6 merely disclose standard crosslinkers (ethylene glycol dimethacrylate, divinyl benzene and allyl methacrylate) used in the art (see for

example D4 col. 4, lines 11 to 34 and D2 lines 4 to 14). The patent does not disclose any technical advantage for employing these claimed crosslinkers. Hence, no inventive step resides in the features of these claims.

8.7 ***Claim 7***

Claim 7 relates to an extrudable resin. Essentially, apart from the added subject matter in respect of the refractive index, the extrudable resin is as defined in claims 1 and 2 and further includes 5 to 50% modifiers. Thus our comments at §8.2 and §8.3 apply equally.

8.7.1 in view of D4 in combination with D2, D6, D10, D7, D11 and D12.

D4 discloses all of the features of claim 7 apart from the feature of 5 to 50% modifiers (see § 7.4.1). However, D2, D6, D10, D7, D11 and D12 teach that it is standard practice in the art to include such modifiers (see D2, p9, lines 27 to 31, D10 col. 3 line 20 onwards and col. 5 lines 11 to 19, D6 col. 6, lines 4 to 19 and D7, D11 and D12) which demonstrate it is common general knowledge to employ modifiers in acrylics.

8.8 ***Claims 8 and 9***

Claims 8 and 9 lack an inventive step for the same reasons as outlined at §8.6.

8.9 ***Claim 10***

It is common practice to include a colorant in resins, particularly a particulate dispersed within a resin (see D6, col. 6, lines 11 to 18 and D9). Thus the subject matter of claim 10 lacks an inventive step.

8.10 ***Claim 11***

Claim 11 relates to an extrudable resin. Essentially, apart from the added subject matter in respect of the refractive index, the extrudable resin comprises the resin as defined in claims 1 and 4. Thus our comments in respect of claims 1 and 4 at §8.2 and §8.5 apply equally. Thus the subject matter of claim 11 lacks an inventive step.

8.11 **Claims 12 and 13**

As explained at §8.2, D1 to D4 relate to producing an article having a frosted and surface textured finish. Thus the subject matter of claims 12 and 13 lack an inventive step.

8.12 **Claim 14**

D1 to D4 all relate to producing articles for lighting, amongst other things, thus the subject matter of claim 14 lacks an inventive step for reasons details before.

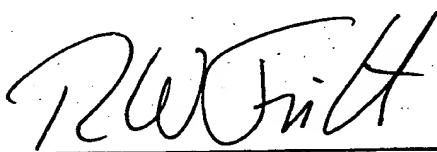
9. **INSUFFICIENCY**

9.1. Should the patentee argue that the above disclosures are inoperative for any reason, then we would argue that the patent is inoperative for the same reason as it fails to disclose any further information.

10. **CONCLUSION**

10.1 In view of the above comments and evidence it is clear that the claims of the patent should be revoked in their entirety.

Signed



Name R W Frith Authorised Representative for
Lucite International UK Limited

✉ EPA/EPO/OEB
D-80298 München
✉ +49 89 2399 - 0
FAX +49 89 2399 - 4465

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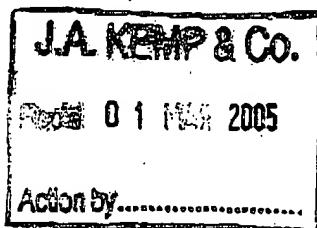
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Direction Générale 2

Marshall, Monica Anne
J.A. KEMP & CO.
14 South Square
Gray's Inn
London WC1R 5JJ
GRANDE BRETAGNE



Formalities Officer

Name: Geeta L.

Tel.: 2568

Date

25-02-2005

Reference N.78777 MAM	Application No./Patent No. 00300413.2 - 2307 / 1022115
Applicant/Proprietor ARKEMA	

Communication of notices of opposition (R. 57(1) EPC)

Notices of opposition have been filed within the opposition period by:

01. Lucite International UK Limited/Queens Gate, 15-17 Queens Terrace/Southampton, Hampshire SO14 3 BP/GB//
02. Röhm GmbH/Kirschenallee/64293 Darmstadt/DE//

The notices of opposition indicated above have been already communicated to you.

You are requested to file your observations within a period of 4 months from notification of this communication.

You may also file amendments, where appropriate, to the description, claims and drawings within the period specified. One set of these documents is to be filed.

If you introduce documents which have not yet been mentioned during the proceedings, your attention is drawn to Rule 59 EPC.

Enclosures:

Opposition Division.



From

Luderschmidt, Schüler & Partners, European Patent Attorneys, Wiesbaden

To the

European Patent Office, Munich, referenced R 664 and dated 23 December 2004

European Patent 1 022 115 (00 300 413.2)

Patentee: Atofina

Opponent: Röhm GmbH & Co. KG

“Polymeric articles having a textured surface and frosted appearance”

We hereby give notice pursuant to Article 99(1) EPC in the name and on behalf of

Röhm GmbH & Co. KG

Kirschenallee

64 293 Darmstadt

Germany

of

OPPOSITION

to the above-identified European patent and request that the granted patent be revoked in its entirety because the subject-matter of the European patent under Articles 52 to 57 is not patentable (Article 100 a) EPC).

The opposition is further supported on the ground that the European patent does not disclose the invention with sufficient clarity or completeness for it to be able to be performed by a person skilled in the art (Article 100 b) EPC).

A further ground for the opposition is that the subject-matter of the European patent goes beyond the content of the application as filed (Article 100 c) EPC).

Oral proceedings are requested should it not be possible for the challenged patent to be revoked in the scope indicated solely on the basis of the written arguments.

The submission of a power of attorney is considered not to be necessary at the present time.

The due opposition fee of EUR 610.00 is paid with the attached debit order.

The grounds are set out on pages attached separately.

For the opponent:

[signature]

Dr. Mai
European Patent Attorney
Association No. 141

Enclosures

- Grounds (2 copies)
- Publications (2 copies)
- Debit order

Grounds**I. Publications**

The opposition is supported on the following prior art:

- D1 Brandrup, J.; Immergut, E.H.; Grulke, E.A. "Polymer Handbook" 1975, pp. III-241 - III-244
- D2 Our own calculations of the difference in refractive index
- D3 DE 35 28 165 A1
- D4 Schildknecht, C.E. "Polymerization processes" New York, London, Sydney, Toronto, Wiley & Sons (1977) pp. 106-142
- D5 JP 61-078859 (Abstract)
- D6 JP 61-159440 (Abstract)
- D7 Saechting "Kunststoff-Taschenbuch" Munich, Vienna, Hanser, 26th Edition (1995), pp. 428-429

II. Subject-matter of the opposed patent

The opposed patent relates according to a first aspect (Claim 1) in the granted version) to

- a) an extruded polymeric article comprising
- b) a polymeric matrix and
- c) polymeric particles
- c-1) which are substantially spherical and
- c-2) highly crosslinked and
- c-3) have a mean particle size of 25-55 μm and
- c-4) a particle size distribution between 10 and 110 μm ,
- a-1) wherein the article has a frosted and textured surface finish,
- a-2) wherein the frosted appearance is achieved through the mismatch (mutual offset) of the refractive indices of the polymeric particles and polymeric matrix by greater than 0.02, and
- b-1) wherein the polymer used as the polymeric matrix is an acrylic polymer and
- c-5) the polymeric particles comprise 10-50% styrene, 90-50% methyl methacrylate and 0.1-2.5% crosslinker (crosslinking agent).

According to a second aspect the opposed patent relates (Claim 7 in the granted version) to an extrudable resin comprising

- a) 20-90% polymethyl methacrylate-based matrix;
- b) 5-50% modifiers; and
- c) 5-30% highly crosslinked spherical beads comprising
10-50% styrene;
90-50% methyl methacrylate; and
0.1-2.5% crosslinking agent,
wherein the beads have a mean diameter of 25-55 μm and a particle size distribution of between 10 and 110 μm , wherein there is a mismatch (mutual offset) of the refractive indices of the crosslinked spherical beads and the polymethyl methacrylate-based matrix by greater than 0.2.

According to a third aspect the opposed patent relates (Claim 11 in the granted version) to an extrudable resin comprising

- a) 70-85% polymethyl methacrylate-based matrix;
- b) 15-30% highly crosslinked spherical beads comprising
15-35% styrene;
65-85% methyl methacrylate; and
0.5-1.0% allyl [sic: cf the german B1 claims] methacrylate,
wherein the beads have a mean diameter of 25-55 μm and a particle size distribution of between 10 and 110 μm , wherein there is a mismatch (mutual offset) of the refractive indices of the crosslinked spherical beads and the polymethyl methacrylate based matrix by greater than 0.2.

According to a fourth aspect the opposed patent relates (Claim 12 in the granted version) to the use of a polymeric matrix and polymeric particles as defined above, for preparing an article having a frosted and textured surface finish.

According to a fifth aspect the opposed patent relates (Claim 13 in the granted version) to the use of a resin as defined in accordance with the second and third aspects for preparing an article having a frosted and textured surface finish.

According to a sixth aspect the opposed patent relates (Claim 14 in the granted version) to the use of an article as defined in accordance with the first aspect, or of a resin as defined in accordance with the second or third aspects, for preparing parts for lighting, signs, point of purchase or cosmetic displays, containers, home or office decorations, furniture applications, shower doors and office doors.

III. Inadmissible extension

First of all it must be observed that the opposed patent is to be revoked pursuant to Article 100 c) EPC. The subject-matter of the patent goes beyond the content of the application as filed.

According to granted Claim 7 the extrudable resin features a polymethyl methacrylate-based matrix and highly crosslinked spherical beads, the mismatch (mutual offset) of the refractive indices of the highly crosslinked spherical beads and the polymethyl methacrylate-based matrix being intended to be greater than 0.2.

In a similar way, the extrudable resin according to Claim 11 features a polymethyl methacrylate-based matrix and highly crosslinked spherical beads, the mismatch (mutual offset) of the refractive indices of the highly crosslinked spherical beads and the polymethyl methacrylate-based matrix being intended to be greater than 0.2.

These claims were submitted together with further amendments on 24 April 2002 in the examination proceedings, and are not supported by the documents as originally filed. In particular the feature "mismatch of greater than 0.2" finds no support in the documents as filed. Those documents disclose only a mismatch (n) of > 0.02 (see page 3, line 28 and page 10, line 7).

Along with Claims 7 and 11, the claims which refer to these claims, namely Claims 8-10, 12, 13 and 14 (in part), fall too.

IV. Insufficient disclosure

Furthermore, the opposed patent is to be revoked pursuant to Article 100 b) EPC. The teaching of the patent is not disclosed with sufficient clarity and completeness that it can be performed by a person skilled in the art.

According to the present Claims 7 and 11 the extrudable resins are to have a mismatch (mutual offset) of the refractive indices of the highly crosslinked spherical beads and of the polymethyl methacrylate-based matrix of more than 0.2. These highly crosslinked spherical beads are to comprise 10-50% styrene, 90-50% methyl methacrylate and 0.1-2.5% crosslinking agent. Using a polymethyl methacrylate-based matrix and spherical beads having such a composition, however, it is no longer possible to achieve a mismatch of more than 0.2, as can be seen, for

example, from the refractive indices of pure polystyrene (D1: 1.59-1.592) and a pure polymethyl methacrylate (D1: 1.4893-1.490).

For further details, we refer to the attached calculations (D2).

Moreover, because of missing units in relation to the percentages, the skilled person is not in a position to determine, even approximately, the range of protection of the present patent. The figures could refer, for example, to % by weight, % by volume, molar %, or to any other units.

V. Novelty

Moreover, it can be assumed that the subject-matter of the present independent claims - in so far as it is defined at all - is not new and hence is not patentable pursuant to Article 52(1) in conjunction with Article 54(2) EPC. The subject-matter claimed has long been known in the art.

Publication DE 35 28 165 A1 (= D3) discloses clouded plastics elements (= feature a)) comprising a polymeric matrix (= feature b)), for example an acrylic resin (see page 5, line 9; = feature b-1), and 3-30% by weight, based on the polymer matrix, of crosslinked beads (= features c), c-1) and c-2)), the crosslinked beads being composed of a polymer synthesized 9.9%-59.9% by weight of one or more free-radically polymerizable monomers which include an aromatic radical, or nonaromatic monomers which include halogen, such as, for example, styrene (see p. 3, l. 48) and 90%-40% by weight of vinylic monomers different from but copolymerizable with the said monomers, such as, for example, methyl methacrylate (see p. 3, ll. 59-65) and 0.1%-20% by weight of at least one crosslinking monomer, such as, for example, allyl methacrylate (see p. 4, ll. 15-16), ethylene glycol dimethacrylate (see p. 4, l. 11) and divinylbenzene (see p. 4, l. 10) and 0%-10% by weight of a strongly polar monomer (= feature c-5)), with the provisos that

- a) the refractive index of the polymer is higher than that of the polymeric matrix, and
- b) the average bead size of the beads is 20-50 μm , preferably 20-35 μm (see page 3, lines 17-29; = feature c-3)). The moulding compound is produced by extrusion (see p. 5, ll. 15-17).

The difference in the refractive indices of the beads and of the polymer matrix is to

have a value of at least 0.02, preferably 0.04 (see p. 5, II. 6-7; = feature a-2)).

On the basis of the composition and the manner of their production the clouded plastics elements automatically have a frosted and textured surface finish (= feature a-1)).

The crosslinked beads are produced in D3 by suspension polymerization, in other words with the same process as in the present opposed patent (see p. 4, II. 20-66). The standard deviation of the particle size of such suspension polymerization is in the majority of cases 20% to 30% (D4: p. 125, II. 19-20), and accordingly the beads possess a particle size distribution in the required range between 10 and 110 μm (feature c-4)).

The disclosure content of publication D3 therefore anticipates at least the subject-matter of Claims 1, 5, 6 and 12 in a manner prejudicial to novelty.

VI. Inventive step

In the opinion of the present party the subject-matter of the opposed patent according to the present independent Claims 1, 5, 6 and 12 has already been described in its entirety by the prior art.

It is also suggested by further publications:

Publication JP 61-078859 (= D5) discloses acrylic resin compositions for lamp covers with high light transmission and high light scattering. The acrylic resin compositions comprise 1 to 20 parts by weight of a crosslinked polymer, for example a polymer having a particle size of 30 μm , which is obtainable by polymerizing a mixture of 10-90% by weight C₁-C₄ alkyl methacrylate, 1-50% by weight of aromatic vinyl monomer and 5-50% by weight of C₁-C₈ alkyl acrylate and 0.3-3% by weight of crosslinking monomer, 0.1 to 5 parts by weight of fine inorganic particles and 100 parts by weight of acrylic resin, polymethyl methacrylate for example.

Since polymers of this kind are prepared by suspension polymerization they also have the particle size distribution required by the opposed patent in the required range between 10 and 110 μm (D4: p. 125, II. 19-20).

polymeric particles and the polymeric matrix, of more than 0.02, comes about automatically in view of the components used.

Publication JP 61-159440 (= D6) discloses acrylic resin compositions for lamp covers with high light transmission and high light scattering. The acrylic resin compositions comprise 1 to 40 parts by weight of a crosslinked polymer, for example a polymer having a particle size of 30 μm , which is obtainable by polymerizing a mixture of 10-90% by weight C₁-C₄ alkyl methacrylate, 20-40% by weight of aromatic vinyl monomer, styrene for example, and 5-50% by weight of C₁-C₈ alkyl acrylate and 0.3-3% by weight of crosslinking monomer and 100 parts by weight of acrylic resin, polymethyl methacrylate for example.

Since polymers of this kind are prepared by suspension polymerization they also have the particle size distribution required by the opposed patent in the required range between 10 and 110 μm (D4: p. 125, II, 19-20).

The mismatch required by the opposed patent between the refractive indices of the polymeric particles and the polymeric matrix, of more than 0.02, comes about automatically in view of the components used.

The subject-matter of Claim 1 according to the opposed patent differs from the disclosure content of the aforementioned publications only in the feature whereby the polymeric particle according to Claim 1 of the opposed patent is obtained by extrusion. This "teaching", however, is trivial in light of the general state of the art, for example the publication D3, and is incapable of substantiating inventiveness.

The same applies to the dependent Claims 2 to 4. The subject-matter of these purportedly particularly appropriate embodiments is likewise suggested by a combination of publications D3, D5 and D6.

It should also be borne in mind that the impact modification of acrylate moulding compounds (see opposed patent: e.g. Claim 2) by adding corresponding modifiers has already long been known in the state of the art (see D7). Exemplary modifiers comprise styrene-modified acrylate elastomers, which are added with an amount of up to 30% by weight and are harmonized, where appropriate, in terms of refractive index.

Claim 14 of the opposed patent describes commonplace fields of application for the

extruded, polymeric articles and resins of the opposed patent. No particular inventiveness can be discerned therein.

In view of all of the above, the requests made at the outset are well founded.

For the opponent

[signature]

Dr. Mai

(No. 141)

D2

R 664 mai/tb

17 December 2004

2004150380

European Patent 1 022 115 (00 300 413.2)**Patentee: Atofina****Opponent: Röhm GmbH & Co. KG****"Polymeric articles having a textured surface and frosted appearance"**

Document D2: Calculation of the difference in refractive index

The difference in refractive index was calculated on the basis of the Gladstone-Dale relationship, assuming additivity of the individual phases (see D1). The contribution of the crosslinker, present according to the opposed patent in amounts of 0.1% to 2.5%, can be disregarded in this context. Besides the pure polymethyl methacrylate matrix, exotic matrices with an extremely high or an extremely low refractive index n were also taken into account, by way of comparison.

Table 1: Calculation of the difference in refractive index

Styrene [wt. %]	MMA [wt. %]	N	Δn^{PMMA}	Δn^{PPDFOA}	Δn^{PNMA}
10	90	1.500	0.010	0.162	-0.141
20	80	1.510	0.020	0.172	-0.131
30	70	1.521	0.031	0.183	-0.120
40	60	1.531	0.041	0.193	-0.110
50	50	1.541	0.051	0.203	-0.100

Styrene: styrene fraction in the polymer

MMA: methyl methacrylate fraction in the polymer

n: calculated refractive index

 Δn^{PMMA} : difference in refractive index from PMMA ($n = 1.49$) Δn^{PPDFOA} : difference in refractive index from poly(pentadecafluoroctyl acrylate) ($n = 1.338$) Δn^{PNMA} : difference in refractive index from poly(naphthyl methacrylate) ($n = 1.641$)

R 664 mai/tb

17. Dezember 2004

2004150380

Europäisches Patent 1 022 115 (00 300 413.2)

Patentinhaber: Atofina

Einsprechende: Röhm GmbH & Co. KG

"Polymeric articles having a textured surface and frosted appearance"

Dokument D2: Berechnung des Brechungsindexunterschiedes

Die Berechnung des Brechungsindexunterschiedes erfolgte auf der Grundlage der Gladstone-Dale Beziehung unter Annahme einer Additivität der einzelnen Phasen (s. D1). Der Beitrag des Vernetzers, gemäß Streitpatent in Mengen von 0,1 % bis 2,5 % vorliegt, kann in diesem Zusammenhang vernachlässigt werden. Neben der reinen Polymethylmethacrylat-Matrix wurden auch vergleichsweise exotische Matrices mit einem extrem hohen oder einem extrem niedrigen Brechungsindex n berücksichtigt.

Tabelle 1: Berechnung des Brechungsindexunterschiedes

Styrol [Gew.-%]	MMA [Gew.-%]	N	Δn^{PMMA}	Δn^{PPFOA}	Δn^{PNMA}
10	90	1,500	0,010	0,162	-0,141
20	80	1,510	0,020	0,172	-0,131
30	70	1,521	0,031	0,183	-0,120
40	60	1,531	0,041	0,193	-0,110
50	50	1,541	0,051	0,203	-0,100

Styrol: Styrolanteil im Polymer

MMA: Methylmethacrylatanteil im Polymer

n: berechneter Brechungsindex

Δn^{PMMA} : Brechungsindexunterschied zu PMMA ($n = 1,49$)

Δn^{PPFOA} : Brechungsindexunterschied zu Poly-(pentadecafluoroctylacrylat)

($n = 1,338$)

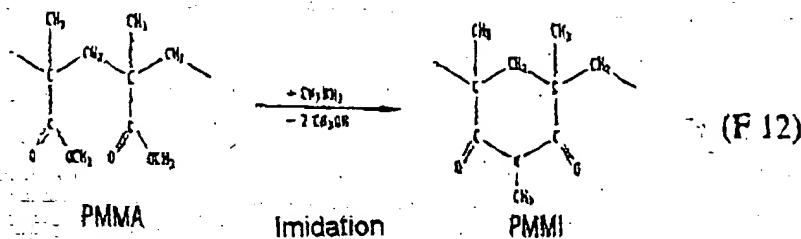
Δn^{PNMA} : Brechungsindexunterschied zu Poly-(-naphthylmethacrylat) ($n = 1,641$)

Fields of use: Bottles for cosmetic products, spray compositions and cleaning compositions; industrial hollow articles; single-use medical devices, fittings and packaging components.

Trade names e.g.: Cyrolite.

4.5.2.3 Polymethacrylimide copolymers, PMMI

PMMI is formally a copolymer of methyl methacrylate (MMA) and glutaramide, but is prepared by reacting PMMA with methylamine (MA) at high temperature and high pressure, see F 12.



Like PMMA it is glass-clear and colourless, has a high light transmittance and no turbidity. The ring closure produces higher chain rigidity and hence a higher heat distortion resistance. Depending on the degree of imidization it is possible to achieve all intermediate values of properties in comparison to PMMA. PMMI has a low oxygen permeability and is less sensitive to stress cracking with respect to ethanol, ethanol/water mixtures and isoctane/toluene mixtures.

Processing by injection moulding takes place, after the granules have been dried initially at 140°C, at melt temperatures of 200 to 310°C and mould temperatures of 120 to 150°C.

Comparison of properties see Table 4.22

Fields of use: Headlamp cover plates for vehicles, street lamp covers, as a blend component and as a fibre-reinforced construction material. For PMMI foams see section 4.10.2.4.

Trade names e.g.: Pleximid, Kamax, PMI-resin

4.5.2.4 High-impact PMMA

High-impact moulding compounds are prepared by suspension or emulsion polymerization in two-phase form. In the PMMA matrix, acrylate elastomers modified, for example, with styrene ($\leq 30\%$, harmonized in terms of refractive index form the toughening disperse phase. As a result of this construction, moulded materials comprising the high-impact compounds, which are of unrestricted miscibility with PMMA, are glass-clear and stable to weathering in the same way as pure PMMA. Their sensitivity to stress cracking is lower and their hot water resistance is better.

Processing takes place after initial drying or with degassing screws by injection moulding and by extrusion at melt temperatures of 210 to 230°C and mould temperatures of 60 to 80°C.

Comparison of properties see Table 4.22

Fields of use: Weathering protection layer in the case of construction profiles such as, for example, PVC window profiles, household appliances, drawing and writing implements; sanitary components, lamp covers.

Trade names e.g.: Diakon, Lucryl, Oroglass, Plexiglas.

4.5.2.5 PMMA + ABS

A blend of this kind is employed for automotive components (housings, reflectors) and also in apparatus construction and in the electrical industry. The material is metallizable, displays good welding characteristics and possesses a better weathering stability and rigidity than ABS.

Trade name: Plexalloy

4.5.2.6 Specialty products, polyacrylate resins

(Trade names in brackets)

Homopolymeric acrylic esters (PAA) are flexible resins whose significance, owing to their good stability to light, oxidative influences and heat, and their elasticizing action, lies in copolymerization and terpolymerization with PS, PVC, VA, MA, AN and acrylic acid. They are supplied as solid resins or solutions, but principally as dispersions (Acronal, Acrysol, Plexigum, Plexisol). *Oxalidine-modified acrylic varnish resins* (Acryloid) are crosslinkable with isocyanates. *Elastoplastic copolymers* are base materials for joint sealers, with those containing > 20% acrylic acid being water-soluble. *Polyhydroxyethyl methacrylate* (Hydron) is used, saturated with about 40% water, for contact lenses and for coating (e.g. of spectacles) and for encasements with controlled water absorptions and water permeabilities in medicine and industry. *Digestive tract-soluble acrylic resins* (Endragit [sic]) are needed for coating medicinal products. By copolymerizing components which are crosslinkable with one another or with secondary components (e.g. isocyanates), heat-curable or radiation-curable film-forming resins are prepared (e.g. Acryplex, Degalan, Larodur, Macrynal, Plex, Scopacron, Synthacryl). *Methacrylate copolymers* formulated for hardness are used as a finishing coat for synthetic leather and other fuel-resistant coatings. *MMA-VC copolymers* (e.g. Paraloid) are elasticizers for PVC. *Unsaturated aliphatic polyurethane-acrylate resins* (Crestomer) are tough, flexible, GRP laminating resins which are crosslinkable with H₂O₂.

4.6 Polyacetals, POM

4.6.1 General description

POM (polyoxymethylene, polyformaldehyde, polyacetals) are partially crystalline thermoplastics which are formed by homopolymerization (POM-H) or copolymerization (POM-

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R) of formaldehyde, see F 13.